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## Evolution of mixing state of black carbon particles: Aircraft measurements over the western Pacific in March 2004

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[1] We report the evolution of the mixing state of black carbon (BC) particles in urban plumes measured by an airborne single particle soot photometer. The aircraft observations were conducted over the ocean near the coast of Japan in March 2004. The number fraction of coated BC particles with a core diameter of 180 nm increased from 0.35 to 0.63 within 12 hours (h), namely  $2.3\% \text{ h}^{-1}$ , after being emitted from the Nagoya urban area in Japan. BC particles with a core diameter of 250 nm increased at the slower rate of  $1.0\% \text{ h}^{-1}$ . The increase in coated BC particles was associated with increases in non-sea salt sulfate and water-soluble organic carbon by a factor of approximately two, indicating that these compounds contributed to the coating on the BC particles. These results give direct evidence that BC particles become internally mixed on a time scale of 12 h in urban plumes. **Citation:** Moteki, N., Y. Kondo, Y. Miyazaki, N. Takegawa, Y. Komazaki, G. Kurata, T. Shirai, D. R. Blake, T. Miyakawa, and M. Koike (2007), Evolution of mixing state of black carbon particles: Aircraft measurements over the western Pacific in March 2004, *Geophys. Res. Lett.*, 34, L11803, doi:10.1029/2006GL028943.

### 1. Introduction

[2] Black carbon (BC) aerosol is emitted from fossil fuel combustion and biomass burning. It efficiently absorbs visible sunlight and heats the atmosphere. The mass-normalized absorption cross section of BC depends on size distribution and mixing state and can increase by factors of  $\sim 1.5$  if BC is coated by non-refractory and non-light absorbing compounds, including sulfate, nitrate, and organics [Bond *et al.*, 2006]. The surface of freshly-emitted BC is coated by non-refractory compounds (e.g., lubricant oil, sulfate) to some extent [Johnson *et al.*, 2005, and references therein] and becomes further coated by secondarily-formed compounds

through the process of gas-to-particle conversion (e.g., condensation). BC, when internally mixed with hygroscopic compounds, acts as cloud condensation nuclei (CCN) and is removed by wet deposition. The  $e$ -folding time of the transformation of BC from hydrophobic to hydrophilic is assumed to be about 1 day in a number of global models [Park *et al.*, 2005, and references therein], but this is without direct evidence based on observations of the mixing states of BC. Therefore, it is critically important to understand the evolution of the mixing state of freshly emitted BC for accurate estimates of the heating rates by individual BC particles and spatial distributions of BC. The mixing states of BC near emission sources and remote areas have been investigated in a number of studies [Clarke *et al.*, 2004, and references therein]. However, the time scale of the changes in the mixing state has not been investigated thus far. This study reports the evolution of the mixing state of BC in urban plumes transported in the marine boundary layer (MBL).

### 2. Aircraft Sampling and Analysis

[3] Aircraft observations of aerosols and trace gases were made over the Pacific Ocean offshore of the Nagoya Urban Area, Japan (NUA;  $\sim 35^\circ\text{N}$ ,  $137^\circ\text{E}$ ) during the Pacific Exploration of Asian Continental Emission-C (PEACE-C) aircraft observation campaign from March 22 to 27, 2004 (Flight 1–5). Aerosol particles were sampled nearly isokinetically ( $\sim 140 \text{ m s}^{-1}$ ) through a stainless steel tube with an inner diameter of 5 mm mounted at the top of the fuselage. The air speed inside the main inlet was gradually slowed down to near the pumping speed of each instrument ( $\sim 2\text{--}6 \text{ m s}^{-1}$ ) by gradually increasing the tube's inner diameter up to 21 mm. Each instrument sampled air from the main inlet through a 3.8–10 mm inner diameter tube. The transmission efficiency of the whole aerosol inlet system was estimated to be close to unity for particles with diameters ( $D_p$ )  $< 1 \mu\text{m}$ , 50% for  $D_p = 1.7 \mu\text{m}$ , and 7% for  $D_p = 2.5 \mu\text{m}$ , based on aerodynamic calculations. The Single-particle soot photometer (SP2) (Droplet Measurement Technologies, Inc., Boulder, CO), which implements the laser-induced incandescence (LII) technique, was used to measure the size and mixing state of BC particles. Details of the SP2 instrument have been described previously [e.g., Baumgardner *et al.*, 2004; Schwarz *et al.*, 2006]. In brief, the SP2 detects LII and scattering signals for individual BC particles. The BC measured by the SP2 is defined as particles that incandesce at a temperature similar to that of laboratory-generated refractory carbon particles (e.g., graphite, glassy carbon, flame-generated soot), and can be discriminated from other light-absorbing aerosols (i.e., non-refractory light absorbing carbon, metals) by the color

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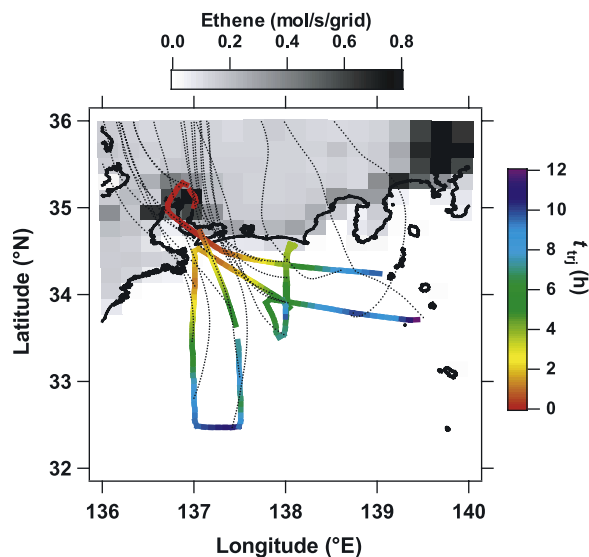
temperatures of incandescence [Schwarz *et al.*, 2006]. The BC mass was estimated from the peak height of the LII signal. The LII peak to BC mass relationship was measured using ambient BC in the laboratory to calibrate the instrument. For this purpose, masses of individual BC particles in ambient air were pre-selected by a system consisting of a heated inlet (400°C), Differential Mobility Analyzer (DMA), and Aerosol Particle Mass analyzer (APM) [Ehara *et al.*, 1996] prior to the detection of the LII signals by SP2. The heated inlet (HI) used in this study is the same as that described by Kondo *et al.* [2006]. They pointed out that the majority (~95%) of non-refractory components (i.e., those which vaporize at  $T < 600^\circ\text{C}$ ) of ambient aerosol (e.g., sulfate, nitrate, organics) are removed by the HI maintained at 400°C. The peak height of the LII signal was shown to be independent of the thickness of coating by non-refractory compounds on BC particles [Schwarz *et al.*, 2006; Moteki and Kondo, 2007]. For this study, BC particle diameter ( $D_{\text{BC}}$ ) was defined as mass equivalent diameter assuming the inherent material density of  $1.77 (\pm 0.07) \text{ g cm}^{-3}$  [Park *et al.*, 2004]. Masses of individual BC particles with  $150 \text{ nm} < D_{\text{BC}} < 800 \text{ nm}$  were measured using the observed LII signals. The precision of the determination of  $D_{\text{BC}}$  was estimated to be 13%. The uncertainty was mainly due to the scattering of the distribution of incandescence intensity for mono-dispersed BC particles. The uncertainty of the BC particle density was assumed to be about 4% ( $\pm 0.07 \text{ g cm}^{-3}$ ) [Park *et al.*, 2004], making a minor contribution to the uncertainty in  $D_{\text{BC}}$ . The detection efficiency of BC by the SP2 was close to unity at  $D_{\text{BC}} > 170 \text{ nm}$  and decreased to about 0.9 at  $D_{\text{BC}} = 150 \text{ nm}$ . A correction was made by taking this efficiency into account for  $150 < D_{\text{BC}} < 170 \text{ nm}$ . The time delay ( $\Delta t$ ) between the scattering peak and LII peak measured by the SP2 depends on the thickness of the BC coating [Schwarz *et al.*, 2006]. With increasing coating thickness,  $\Delta t$  was found to increase abruptly from about  $1 \mu\text{s}$  to about  $4 \mu\text{s}$  at the threshold  $D_p/D_{\text{BC}}$  ratio of about 2 [Moteki and Kondo, 2007]. Here,  $D_p$  denotes the particle diameters of the coated BC particles. According to the observed  $\Delta t$ , we classified the coating condition of BC into two categories, thinly-coated ( $\Delta t < 2 \mu\text{s}$ ) and thickly-coated ( $\Delta t > 2 \mu\text{s}$ ). The  $D_p/D_{\text{BC}}$  ratio is larger than about 2 for thickly-coated BC and smaller than about 2 for thinly-coated BC. The uncertainty in the threshold  $D_p$  is estimated to be about 50% of the  $D_{\text{BC}}$  (i.e.,  $D_p/D_{\text{BC}}$  of  $2.0 \pm 0.5$ ) based on laboratory experiments [Moteki and Kondo, 2007]. Aerosol particles sampled by the SP2 were dry because dried air was used as sheath flow for sample aerosol. Water soluble organic carbon (WSOC) was measured by a system combining the Particle-Into-Liquid Sampler with a total organic carbon (TOC) analyzer (PILS-WSOC) [Sullivan *et al.*, 2004] with an integration time of 6 min. Water soluble inorganic ions ( $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , etc.) were measured by PILS with Ion Chromatography analysis (PILS-IC) [Weber *et al.*, 2001] with an integration time of 5 min. Performances of the PILS-WSOC and PILS-IC systems used in this study are described by Miyazaki *et al.* [2006] and Takegawa *et al.* [2005], respectively. The absorption coefficient of refractory aerosol ( $B_{\text{abs}}$ ) was measured by a Particle Soot Absorption Photometer (PSAP; Radiance Research, Inc.) with a HI maintained at a temperature of 400°C (HI-PSAP). Non-refractory aerosol particles

and coatings on BC were removed by the HI as discussed above. We assumed that mineral dust aerosols did not contribute to  $B_{\text{abs}}$  because  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  concentrations were below the limit of detection ( $< 0.1 \mu\text{g m}^{-3}$ ). We also assumed that only BC without coating contributed to the  $B_{\text{abs}}$ . The mass concentration of BC was derived from  $B_{\text{abs}}$  assuming a mass absorption coefficient (MAC) =  $8.9 \text{ m}^2 \text{ g}^{-1}$ , after the correction of  $B_{\text{abs}}$  based on the procedure described by Bond *et al.* [1999]. The MAC was determined by comparison between  $B_{\text{abs}}$  measured by HI-PSAP with the mass concentration of elemental carbon (EC) measured in Tokyo [Kondo *et al.*, 2006]. In the present study, the uncertainty of the MAC is estimated to be 22%, which is the uncertainty of the EC measurement [Kondo *et al.*, 2006]. Mixing ratios of non-methane hydrocarbons were measured by a whole air sampling technique described by Colman *et al.* [2001], with sampling intervals of 3–4 minutes. Only ethene ( $\text{C}_2\text{H}_4$ ) and ethyne ( $\text{C}_2\text{H}_2$ ) were used in the present analysis. Concentrations of sulfur dioxide ( $\text{SO}_2$ ) were measured by a pulsed UV fluorescence instrument that was used for the PEACE-A and B aircraft campaigns [Parrish *et al.*, 2004].

### 3. Results and Discussion

#### 3.1. Meteorological Conditions and Plume Ages

[4] Flight 5 of PEACE-C was conducted on March 27 over the region of  $35.3\text{--}32.5^\circ\text{N}$  and  $137\text{--}139.5^\circ\text{E}$ . The altitude range covered was 0.3–4 km. We used only data from flight 5, because they were free from the effects of cloud processing on the observed plumes. The thickness of the MBL was estimated to be about 1 km, based on the altitude profiles of BC and relative humidity (RH). The weather was generally fair and the relative humidity (RH) was about  $50 \pm 10 (1\sigma) \%$  in the MBL. We use only the data obtained in the MBL for the present analysis. Figure 1 shows the flight tracks and backward trajectories. The trajectories were calculated using wind fields calculated by the PSU/NCAR mesoscale model (MM5) driven by Japan Meteorological Agency (JMA) global analysis data. Northerly winds transported the pollution plumes emitted from the NUA over the Pacific Ocean in the MBL. The  $\text{C}_2\text{H}_4/\text{C}_2\text{H}_2$  ratio ( $\text{pptv pptv}^{-1}$ ) was used to estimate the photochemical age ( $t_{\text{age}}$ ) of the plume by using the method described by Takegawa *et al.* [2004]. Here  $\text{C}_2\text{H}_4$  and  $\text{C}_2\text{H}_2$  represent the observed  $\text{C}_2\text{H}_4$  and  $\text{C}_2\text{H}_2$  mixing ratios. The  $t_{\text{age}}$  is controlled by the oxidation of  $\text{C}_2\text{H}_4$  and  $\text{C}_2\text{H}_2$  by the hydroxyl radical (OH). The average OH concentration was assumed to be  $4.5 \times 10^6 \text{ cm}^{-3}$ , which was the daytime median value of OH observed over the Pacific Ocean near Japan ( $25\text{--}40^\circ\text{N}$ ,  $125\text{--}150^\circ\text{E}$ ) below 1 km during the TRACE-P campaign (February–April 2001) [Mauldin *et al.*, 2003]. The  $\text{C}_2\text{H}_4/\text{C}_2\text{H}_2$  ratio at  $t_{\text{age}} = 0$  was fixed at  $1.13 (\pm 0.22)$ , which is the average ( $\pm\sigma$ ) value ( $n = 14$ ) for fresh urban outflow observed during the PEACE-C campaign. This ratio is in agreement with the value  $1.24 (\pm 0.51)$  observed in Tokyo ( $35.4^\circ\text{N}$ ,  $139.4^\circ\text{E}$ ) from January to February in 2004. Figure 1 also shows the emission inventory of  $\text{C}_2\text{H}_4$  compiled by the Japanese Ministry of Environment (JMOE). The emissions of  $\text{C}_2\text{H}_4$  are concentrated in the populated coastal region of Japan. The transport time ( $t_{\text{trj}}$ ) from the coastline was calculated for each trajectory



**Figure 1.** Flight tracks of the aircraft observations and backward trajectories (dotted lines) of observed air masses. Emission rates of  $C_2H_4$  per  $20\text{ km} \times 20\text{ km}$  grid are shown in gray scale. Transport times ( $t_{trj}$ ) of air masses from the coastline of Japan are shown in colored scale.

and they are shown in Figure 1 as a color scale on the flight track. The median (25th/75th percentile) value of  $t_{age}/t_{trj}$  is 1.2 (0.6/2.3) ( $n = 26$ ). The median value of  $t_{age}/t_{trj}$  is close to unity, supporting the use of the  $t_{age}$  as the time elapsed since emission of the plumes. Plumes with  $t_{age}$  ranged from 0 to 16 h were observed.

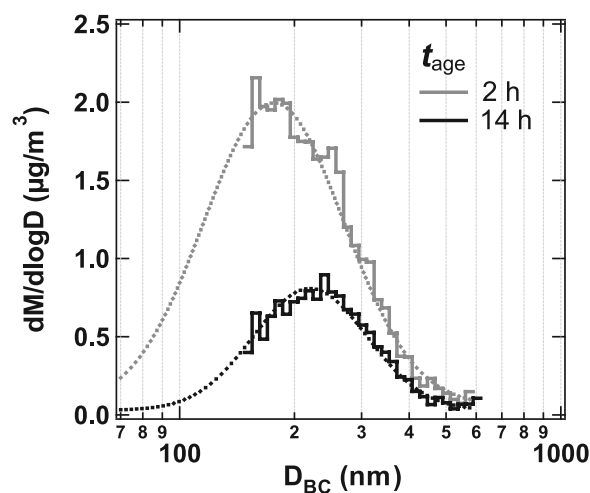
### 3.2. Size Segregated Mixing State of BC

[5] The  $e$ -folding time of air exchange between the MBL and the free troposphere (FT) is estimated to be 45–90 h by using the entrainment velocity range of  $0.3\text{--}0.6\text{ cm s}^{-1}$  [Raes, 1995]. The effects of the FT air on BC observed in the urban plumes should be small because the time scale of mixing is at least 3 times longer than the maximum observed  $t_{age}$ . BC concentrations in the FT were less than  $0.1\text{ }\mu\text{g m}^{-3}$  during flight 5, and the background BC concentrations for the MBL were estimated to be less than  $0.05\text{ }\mu\text{g m}^{-3}$  from the maritime air observed during flights 1–4. These BC concentrations are an order of magnitude lower than those observed in the urban plumes ( $0.5\text{--}1.1\text{ }\mu\text{g m}^{-3}$ ). Therefore, mixing of the plumes with the background air should have little effect on the size distribution and number fraction of thickly-coated BC. The size distributions of BC measured by SP2 for  $t_{age} = 2$  and 14 h are shown in Figure 2. They are fitted with log-normal distributions with mass median diameters (MMD) of 190 and 210 nm (with geometrical standard deviations of 1.55 and 1.45) for  $t_{age} = 2$  and 14 h, respectively. This MMD and width of BC size distribution is similar to those obtained in the lower-to-upper troposphere [Clarke et al., 2004; Schwarz et al., 2006]. Figure 3a shows histograms of the frequency of time delay,  $\Delta t$  for BC observed at different  $t_{age}$ , for BC particles with  $D_{BC} = 180\text{ nm}$ . The frequency distributions of  $\Delta t$  were bimodal and had one population at around  $\Delta t \sim 1\text{ }\mu\text{s}$  (short- $\Delta t$ ) and another at  $\sim 4\text{ }\mu\text{s}$  (long- $\Delta t$ ). The short- $\Delta t$  and the long- $\Delta t$

correspond to thinly-coated BC particles with  $D_p/D_{BC} < \sim 2$  and thickly-coated BC particles with  $D_p/D_{BC} > \sim 2$ , respectively, as discussed in section 2. It is seen that the number fraction of thickly-coated BC was much larger at  $t_{age} = 14\text{ h}$  than that at  $t_{age} = 2\text{ h}$ . Figure 3b shows the changes of the number fraction of thickly-coated BC as a function of  $t_{age}$ . The average ( $\pm\sigma$ ) fraction of thickly-coated BC increased from  $0.35 (\pm 0.05)$  to  $0.63 (\pm 0.09)$  within 12 h. The rate of increase in the thickly-coated number fraction ( $R_{coat}$ ) averaged  $2.3\% \text{ h}^{-1}$ . This increase of the thickly-coated BC is likely due to the oxidation of the precursor gases, followed by condensation of non-refractory compounds, including sulfate and organics, onto the BC particles as discussed in section 3.3. The  $R_{coat}(D_{BC})$  was observed to be lower for the larger  $D_{BC}$ . For example,  $R_{coat}(250\text{ nm}) = 1.0\% \text{ h}^{-1}$  on average.  $R_{coat}(180\text{ nm})/R_{coat}(250\text{ nm})$  was observed to be 2.3. Here we interpret the  $D_{BC}$  dependence of  $R_{coat}(D_{BC})$  using a simple condensation model, which assumes that  $dD_p/dt = A/D_p$ , where  $A$  is a constant [Seinfeld and Pandis, 1998]. If we assume that the initial distribution of  $D_p/D_{BC}$  is independent of  $D_{BC}$ , then  $R_{coat}(180\text{ nm})/R_{coat}(250\text{ nm})$  is calculated to be 1.9, which agrees with the observed ratio to within 20%. The fraction of the thickly-coated BC is a qualitative parameter in defining the changes in the distribution of coating thickness to some extent; however, it is a useful measure in estimating the time scale with which the internal mixing of BC proceeds.

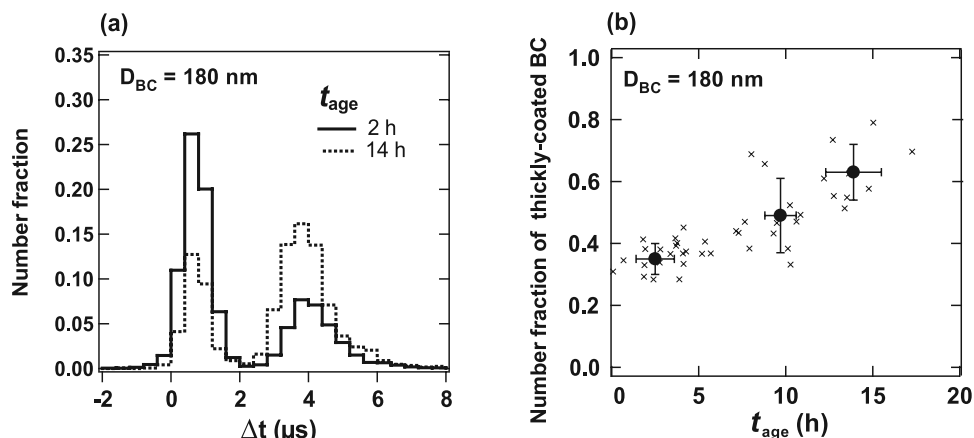
### 3.3. Formation of Non-Refractory Compounds

[6] The evolution of the internal mixing of BC is further interpreted in terms of formation of non-refractory aerosol compounds with increasing  $t_{age}$ . For statistical analysis, the data were classified into three categories with different ranges of  $t_{age}$ : 0–4 (average  $\pm 1\sigma = 2.7 \pm 1.1$ ) h, 8–12 ( $9.8 \pm 0.7$ ) h, and 12–16 ( $13.9 \pm 1.9$ ) h. The BC can be used as a tracer of anthropogenic pollutants for the analysis of the formation of non-refractory aerosol because BC has no



**Figure 2.** Mass size distributions of BC particles measured by SP2 for fresh ( $t_{age} = 2\text{ h}$ ) and aged ( $t_{age} = 14\text{ h}$ ) air masses. The integration time for these size distributions was about 5 min. Log-normal functions were fitted to the data for size ranges of  $150\text{ nm} < D_{BC} < 600\text{ nm}$ .



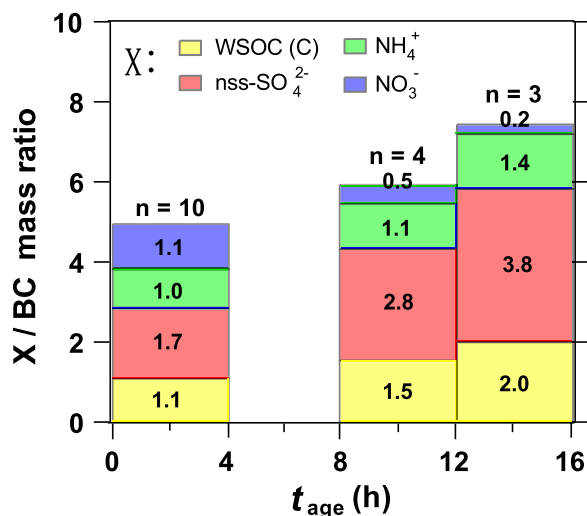


**Figure 3.** (a) Histograms of the time delay ( $\Delta t$ ) of BC particles with  $D_{\text{BC}} = 180 \pm 5 \text{ nm}$  detected by SP2 for photochemical ages ( $t_{\text{age}}$ ) of 2 and 14 h. (b) The number fraction of thickly-coated BC particles ( $\Delta t > 2 \mu\text{s}$ ) versus  $t_{\text{age}}$ . Small crosses indicate data merged with the integration time for the whole air samplings. The filled circles with bars indicate the data averaged over each category of  $t_{\text{age}}$ , as discussed in section 3.3. Vertical and horizontal bars indicate the  $1\sigma$  values.

sources over the ocean. The average  $\pm 1\sigma$  of the BC mass concentrations measured by HI-PSAP for each  $t_{\text{age}}$ -category were  $1.1 \pm 0.2$ ,  $0.8 \pm 0.2$ , and  $0.5 \pm 0.02 \mu\text{g m}^{-3}$ , respectively. The averages ( $\pm 1\sigma$ ) of the  $(\text{SO}_2 + \text{nss-SO}_4^{2-})/\text{BC}$  ratios were  $5.6 \pm 0.7$ ,  $5.1 \pm 0.8$ , and  $5.4 \pm 0.7 \mu\text{g m}^{-3}/\mu\text{g m}^{-3}$  for each  $t_{\text{age}}$ -category, indicating that the air masses originated from areas (depicted in Figure 1) with similar  $\text{SO}_2/\text{BC}$  emission ratios.  $\text{C}_2\text{H}_2$  used to derive  $t_{\text{age}}$  was highly correlated with BC ( $r^2 = 0.72$ ,  $n = 41$ ) for the entire range of  $t_{\text{age}}$  (0–16 h). Considering these results, we assumed that the emission ratios of other precursor gases to BC also varied little over the source regions of the observed plumes for the discussion of secondary aerosol formation.

[7] The increase in the ratios of mass concentrations of aerosol composition X to that of BC ( $\Delta(X/\text{BC})$ ) can be used

as a measure of secondary formation of X. Figure 4 shows the ratios of the mass concentrations of WSOC,  $\text{nss-SO}_4^{2-}$ ,  $\text{NH}_4^+$ , and  $\text{NO}_3^-$  to those of BC for three  $t_{\text{age}}$ -categories. Overall increase in each X/BC were  $\Delta(\text{WSOC}/\text{BC}) = 0.9$ ,  $\Delta(\text{nss-SO}_4^{2-}/\text{BC}) = 2.1$ , and  $\Delta(\text{NH}_4^+/\text{BC}) = 0.4$ . Inorganic ions  $\text{NH}_4^+$ ,  $\text{SO}_4^{2-}$ , and  $\text{NO}_3^-$  were in the forms of ammonium sulfate  $((\text{NH}_4)_2\text{SO}_4)$  and ammonium nitrate  $(\text{NH}_4\text{NO}_3)$  considering that the average ( $\pm 1\sigma$ ) molar ratio of  $\text{NH}_4^+/(2\text{SO}_4^{2-} + \text{NO}_3^-)$  for each  $t_{\text{age}}$ -category were 1.0 ( $\pm 0.3$ ), 1.0 ( $\pm 0.3$ ), and 0.9 ( $\pm 0.1$ ), respectively. From these results, it is likely that ammonium sulfate and secondary organic compounds formed during the transport should have mainly contributed to the increase in the coating on BC particles.



**Figure 4.** BC-normalized mass concentration of aerosol chemical compositions for three categories of the photochemical age ( $t_{\text{age}}$ ):  $t_{\text{age}} = 0\text{--}4$ ,  $8\text{--}12$ , and  $12\text{--}16 \text{ h}$ . The WSOC mass concentrations are in the units of  $\mu\text{gC m}^{-3}$ .

[8] **Acknowledgments.** We are indebted to all the PEACE-C participants for their cooperation. Special thanks are due to Mitsubishi Diamond Air Service Co. We thank S. Hatakeyama for providing the  $\text{SO}_2$  data. We thank D. Baumgardner, N. Oshima, and Y. Morino for giving us useful comments. This work was supported by the Ministry of Education, Culture, Sports, Science, and Technology (MEXT). The Earth Observation Research and Application Center (EORC) of JAXA supported the PEACE campaigns.

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